



## Effects of the injection grout Silica sol on bentonite

Michael Holmboe<sup>a,\*</sup>, Susanna Wold<sup>a</sup>, Torbjörn Petterson<sup>b</sup>

<sup>a</sup> KTH School of Chemical Science and Engineering, Division of Nuclear Chemistry, Royal Institute of Technology, SE-100 44 Stockholm, Sweden

<sup>b</sup> Fibre Technology, Royal Institute of Technology, SE-100 44 Stockholm, Sweden

### ARTICLE INFO

#### Article history:

Available online 31 July 2011

#### Keywords:

Bentonite  
Bentonite colloids  
Silica sol  
Silica colloids  
Colloidal stability  
Colloid interactions

### ABSTRACT

Silica sol, i.e., colloidal SiO<sub>2</sub>, may be used as a low-pH injection grout for very fine fractures in the construction of deep geological repositories for radioactive waste in Sweden and in Finland. If the bentonite barrier encounters SiO<sub>2</sub>-colloid particles under conditions favorable for aggregation, there is concern that it will modify the bentonite barrier at the bentonite/bedrock interface. In this study qualitative experiments were performed with mixed dispersions of SiO<sub>2</sub>-colloids and bentonite or homo-ionic Na/Ca-montmorillonite. Samples were prepared at different colloid concentrations and treated under various conditions such as low and high ionic strength (0.3 M NaCl), as well as dehydration and redispersing. Free swelling and settling experiments were performed in order to qualitatively compare the conditions in which SiO<sub>2</sub>-colloids affect the bulk/macro properties of bentonite. In order to study specific SiO<sub>2</sub>-colloid/montmorillonite interactions and preferred type of initial aggregation, dilute dispersions of homo-ionic montmorillonite dispersions mixed with varying concentrations of SiO<sub>2</sub>-colloids were prepared and selected samples were characterized by PCS, SEM/EDS, AFM and PXRD. The results from this study show that bentonite and montmorillonite particles can be modified by SiO<sub>2</sub>-colloids when mixed in comparable amounts, due to dehydration or high ionic strength. Some indications for increased colloidal stability for the SiO<sub>2</sub>-colloid modified clay particles were also found. From the AFM investigation it was found that initial attachment of the SiO<sub>2</sub>-colloids in Na<sup>+</sup> dominated samples seemed to occur on the edges of the montmorillonite layers. In Ca<sup>2+</sup> dominated samples not subjected to excess NaCl, SiO<sub>2</sub>-colloid sorption onto the faces of the montmorillonite layers was also found. In all, contact between the bentonite barrier and gelled Silica sol should preferably be avoided.

© 2011 Elsevier Ltd. All rights reserved.

### 1. Introduction

During the construction phase of the deep geological repositories for nuclear waste in Sweden and Finland, the hydraulic conductivity of the bedrock must be controlled and kept at a low level which is to be accomplished by the use of suitable injection grouts. A potential candidate for very fine fractures is the gel formed from injected Silica sol (hydrolyzed SiO<sub>2</sub>-colloids), using NaCl as an accelerator. Compared with other conventional cement-based injection grouts, Silica sol has the ability to seal finer fractures with apertures of 10–170 μm, and gives rise to leachate water with pH < 11 (Bodén and Sievänen, 2005; Funehag, 2008; Hölttä, 2008). As many other countries, Sweden and Finland also plan to use compacted bentonite as a barrier in their multi-barrier concepts for storage of nuclear waste. So far the effects and possible interactions of the hydrolyzed Silica sol particles with bentonite under repository conditions have not been well studied. Regardless of how or when the bentonite barrier may be subjected

to SiO<sub>2</sub>-colloids, the effect of deposition of large quantities of hydrolyzed SiO<sub>2</sub> into the immediate vicinity of the bentonite barrier should be investigated. In this study the general effects of SiO<sub>2</sub>-colloids on bulk/macro properties of bentonite is investigated as well as the initial and preferred form of montmorillonite/SiO<sub>2</sub>-colloid aggregation in mixed montmorillonite/SiO<sub>2</sub>-colloid systems.

#### 1.1. Background

During excavation of the host rock Silica sol is planned to be pumped into drilling holes or fractures, directly after mixing with an accelerator composed of 10 weight% NaCl in a 5:1 ratio of SiO<sub>2</sub>-colloids to NaCl. This procedure induces rapid colloid aggregation and gel formation in the boreholes/fractures usually within 1 h and reduces the hydraulic conductivity and groundwater inflow to desired levels (<1 L per minute and 60 m tunnel, Funehag, 2008). Since a full deterministic description of the fracture zones often is very difficult, grouting is always subjected to uncertainties. If the injected SiO<sub>2</sub>-colloids would encounter large water-bearing fractures, dilution with decreasing aggregation rate and increasing gel time is probable. The final extent and methodology of grouting

\* Corresponding author. Tel.: +46 (0)8 7908789; fax: +46 (0)8 7908772.  
E-mail address: [holmboe@kth.se](mailto:holmboe@kth.se) (M. Holmboe).

is never determined *a priori*, but worked out along the way during the construction of the repository (Emmelin et al. 2007).

The probability, however, of the hydrolyzed SiO<sub>2</sub>-colloids encountering the bentonite barrier is considered to be low since most expected grouting operations will take place before deposition of the bentonite barriers. If the bentonite barrier would encounter the hydrolyzed SiO<sub>2</sub>-colloids under conditions favorable for aggregation during the construction phase or later, the expected properties of the bentonite barrier at the bentonite/bedrock interface may be altered. This may affect future colloid generation and bentonite erosion in the event of intrusion of glacial melt water, which is one of the most critical scenarios for the bentonite barrier since mass loss can be substantial under certain conditions (Le Bell, 1978, Laaksoharju and Wold, 2005, Neretnieks et al. 2009). On the other hand, if the gel from Silica sol deposited in the fracture zones proves to be stable towards time and changing groundwater conditions, erosion of bentonite may be inhibited. The long-time behavior and stability of the Silica sol gel itself is not well known, but it is generally assumed that it will be in operation and remain stable for 100 years. Release of the primary hydrolyzed SiO<sub>2</sub>-colloids from the gel is expected to be small (Hölttä et al. 2009), especially at elevated ionic strength. The solubility of the amorphous SiO<sub>2</sub>-particles are expected to be approximately one magnitude higher than quartz (Gunnarsson and Arnórsson, 2000; Karnland et al. 2007). Thus with changing groundwater conditions, the SiO<sub>2</sub> gel would be more likely to undergo solubility-precipitation reactions compared to similar crystalline minerals present in the bedrock.

In general, under low ionic strength groundwater conditions direct aggregation between hydrolyzed SiO<sub>2</sub>-colloids and bentonite particles would not be expected due to the unfavorable charge conditions between the negatively charged particles. However, when subjected to dehydration or elevated ionic strength, especially in the presence of Ca<sup>2+</sup> (Iler, 1979; Lee and Moon, 2004; Kinsela et al. 2010), the SiO<sub>2</sub>-colloids will unavoidably aggregate and coalesce into larger particles and aggregates, either with bentonite or with other SiO<sub>2</sub>-colloids.

Compared with montmorillonite colloids, hydrolyzed SiO<sub>2</sub>-colloids show much higher colloid stability at slightly alkaline conditions, even under relatively high ionic strength (Iler, 1979). Thus, if the hydrolyzed SiO<sub>2</sub>-colloids prove to be reactive towards montmorillonite, an increase in colloid stability of the modified montmorillonite/SiO<sub>2</sub>-colloid particles compared to montmorillonite particles is probable. When aggregating homogeneously, either under high ionic strength or because of dehydration, the hydrolyzed SiO<sub>2</sub>-colloids can form covalent siloxane bonds through the reactive siloxane surface groups (Iler, 1979). In the presence of bentonite, the same type of irreversible reaction may occur along the edges of the montmorillonite layers due to the silanol bonds at the montmorillonite edges. If the hydrolyzed SiO<sub>2</sub>-colloids show a large tendency for heterogeneous aggregation, i.e., react and aggregate irreversibly with the edges of the montmorillonite particles, both the physical/macro properties (swelling pressure, rheology) and the colloidal properties of the Silica-modified montmorillonite particles may be drastically different from those of unmodified montmorillonite. With regards to this, there is concern that SiO<sub>2</sub>-colloids may considerably change the bulk properties and colloidal behavior of bentonite.

In summary, several hypothetical scenarios of the hydrolyzed SiO<sub>2</sub>-colloids encountering the bentonite (or vice versa) exist: (i) Unforeseen SiO<sub>2</sub>-colloid transport due to underestimation of the hydraulic capacity of the fracture zones and inadequate gel formation during grouting. (ii) Transport of bentonite into fractures grouted with SiO<sub>2</sub>-colloids, in the event of bentonite erosion. (iii) Solubility-precipitation reactions of secondary SiO<sub>2</sub>-particles with bentonite.

In this study the general effects of SiO<sub>2</sub>-colloids on the bulk/macro properties of bentonite and more specifically the initial and preferred form of montmorillonite/SiO<sub>2</sub>-colloid attachment in mixed montmorillonite/SiO<sub>2</sub>-colloid systems are investigated. Qualitative bulk/macro experiments and colloid/aggregate characterization of mixed bentonite/SiO<sub>2</sub>-colloid systems has been performed using photon correlation spectroscopy (PCS), atomic force microscopy (AFM), scanning electron microscopy (SEM) and powder X-ray diffraction (PXRD). In order to determine the possible conditions under which aggregation between montmorillonite and SiO<sub>2</sub>-colloids would be possible, different screening aggregation experiments were performed under four conditions: (i) low ionic strength, (ii) high ionic strength, (iii) dehydration (drying), and (iv) high ionic strength and dehydration. Condition (i) was taken as the reference case, since montmorillonite/SiO<sub>2</sub>-colloid aggregation under ambient conditions is not expected. The high ionic strength conditions were set according to the corresponding NaCl concentration (0.3 M) used in injection experiments in Äspö Hard Rock Laboratory, Sweden. Would the bentonite barrier not be fully water-saturated when coming into contact with the SiO<sub>2</sub>-colloids, either due to heat evolving from the nuclear fuel or due to slow water-saturation of the bentonite barrier, aggregation due to dehydration may also be a plausible scenario. This is because the SiO<sub>2</sub>-colloids aggregate irreversibly if subjected to a water content below 35 weight%, according to the manufacturer.

## 2. Material and methods

### 2.1. Materials

Silica sol is a commercially available product with several different applications. As an injection grout, Silica sol has successfully been applied to several tunnel constructions in Sweden. The specific Silica sol used in this study, Meyco MP 320 (EKA Chemicals, Sweden) has been tested as an injection grout *in situ* at Äspö Hard Rock Laboratory, Sweden and in Olkiluoto, Finland. This product consists of hydrolyzed and negatively charged SiO<sub>2</sub>-colloids and has a particle size ranging from 3 to 70 nm, with an average size of approximately 15 nm. The bentonite used in this study was MX-80 Wyoming bentonite with a montmorillonite content of approximately 83%. The exchangeable cations for this bentonite are approximately: 75% Na<sup>+</sup>; 17% Ca<sup>2+</sup>; 7% Mg<sup>2+</sup>; 2% K<sup>+</sup>. The homo-ionic sodium and calcium montmorillonites used were purified MX-80 Wyoming bentonite according to Karnland et al., 2006. The CEC for MX-80 and the purified Na-montmorillonite measured by a modified Cu(II)-trien method is ~0.75 and ~0.85 meq/g, respectively (Karnland et al., 2006).

### 2.2. Method

#### 2.2.1. Free swelling and settling experiments with bentonite

Free swelling and settling experiments were performed in order to qualitatively compare the conditions in which SiO<sub>2</sub>-colloids affect the bulk/macro properties of bentonite. The experiments were performed with mixed bentonite and SiO<sub>2</sub>-colloid dispersions under different conditions representing the four postulated scenarios, (i) low ionic strength, (ii) high ionic strength, (iii) dehydration, and (iv) high ionic strength and dehydration. In the free swelling experiments, 1 g of bentonite was allowed to expand in the presence of varying amounts of SiO<sub>2</sub>-colloids, with or without NaCl addition to a total volume of 100 mL for 2 months. The final volume and appearance of the swelling bentonite-SiO<sub>2</sub>-colloid mixtures were inspected visually and photographed. The samples were first prepared in 10 mL of water with 0; 0.1; 0.5 g SiO<sub>2</sub>-colloids (with or without addition of NaCl(aq)). Water and SiO<sub>2</sub>-colloids were added

Download English Version:

<https://daneshyari.com/en/article/4721236>

Download Persian Version:

<https://daneshyari.com/article/4721236>

[Daneshyari.com](https://daneshyari.com)